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Neutron Scattering of CeNi at the SNS-ORNL: A Preliminary Report

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ABSTRACT

This is a preliminary report of a neutron scattering experiment used to investigate 4f electron behavior in Ce.

INTRODUCTION

The manifestations of electron-correlation in Pu and Ce have interesting parallels [1], including large volume collapses between phases. [2,3] CeNi, using Ni 60 to minimize the Ni nuclear scattering [4], was chosen as an avenue to probe the magnetic cancellation in Ce. This magnetic cancellation should be of the Kondo type, with the valence electrons screening the f electron moment. [5-7] This screening should change under pressure. [8,9] The neutron scattering experiments [10] were carried out at the Spallation Neutron Source [11] at Oak Ridge National Laboratory, using the Sequoia Facility. [12]

CONTEXT WITH RESPECT TO PLUTONIUM

Pu has a longstanding history of being an incredibly important material that has defied analysis. [13-18] Despite great effort and important contributions by many authors [19-30] the nature of the Pu 5f electron remains unclear. It is now widely accepted that some sort of electron correlation [22,23,30] must be responsible for the magnetic cancellation [28-31], but the specifics are not understood. Many possible hypotheses [28] have been advanced, including Kondo-like spin shielding with a multi-configurational approach using Dynamical Mean Field Theory (DMFT) [22] and a magnetic cancellation between the orbital and spin components of the 5f manifold. [30]. What is lacking is a true experimental benchmarking

The importance of resolving the Pu 5f issue is manifold. First, it is one of the last, great, unsolved problems in Condensed Matter Physics. [13-31] Second, there are very important technological, industrial, societal and environmental ramifications. Besides its importance in Defense Applications [18], Pu is a crucial material in energy production [13,14,34] and long-term nuclear waste disposal. [13,14,35] How can we predict the way Pu will behave over the 10,000 year period mandated for storage if we don't even understand its ground state electronic structure?

For example, it is widely known [13,14,23] that Pu has six solid phases, with the most dense being monoclinic, not a high symmetry phase such as face-centered-cubic (fcc). In fact, there is a 25% volume change between the monoclinic α and the fcc δ and to stabilize the less-dense fcc δ-Pu, it is necessary to alloy it with materials such as Ga. [13,14,18,31] Sometimes, the claim is made that there is transition between itinerant and localized 5f behavior between these two phases. Unfortunately, it is not as simple as that and electron correlation is probably playing a large role in all of the Pu5f behavior.

EXPERIMENT and DISCUSSION

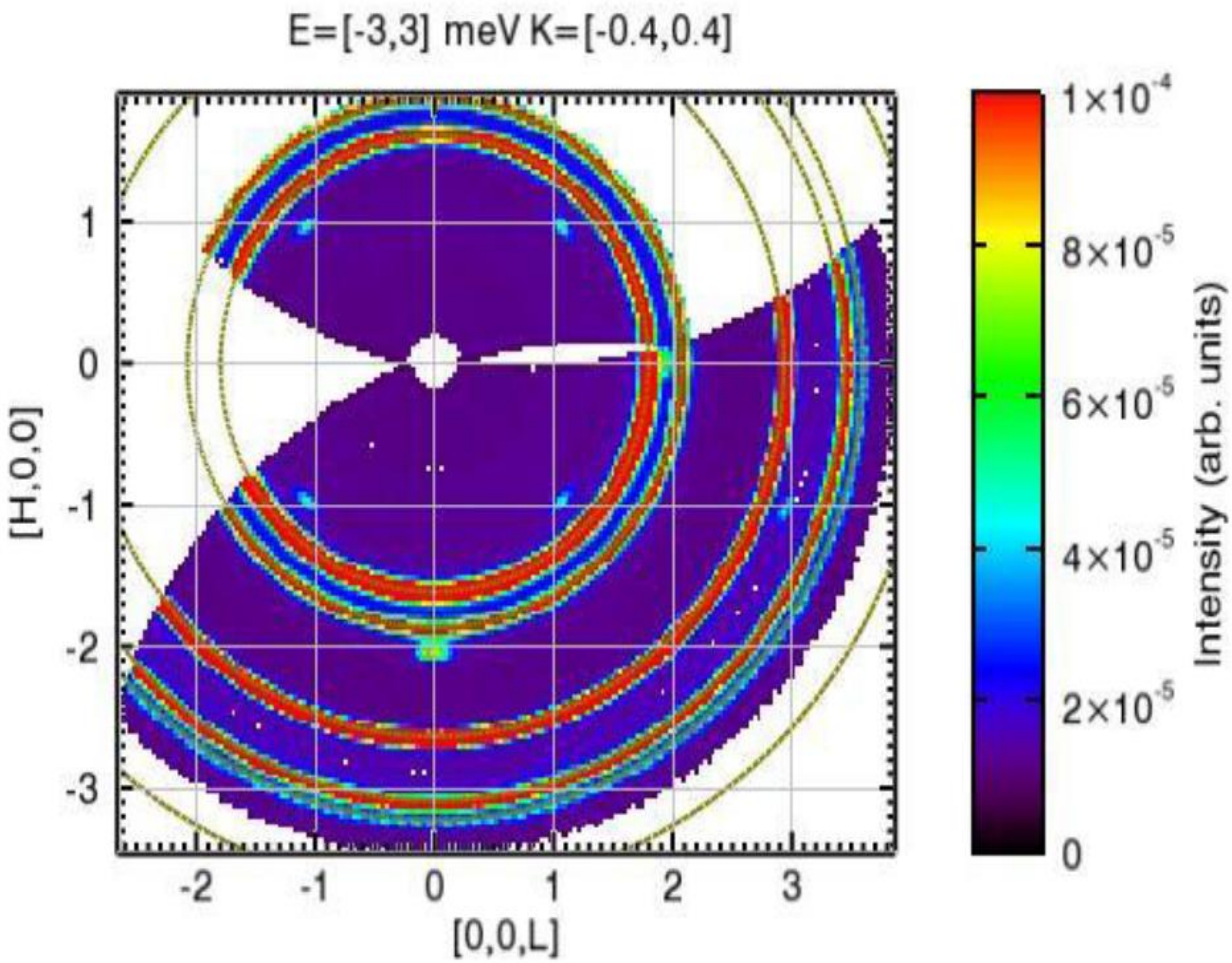

Figure 1: An example of scattering data from CeNi. The pie-slice-shaped sections are derived from rotation of the sample. The distortions away from linear cuts are due to the conversion from angle to momentum.

Pulsed neutrons are generated by the acceleration of protons into a target. The pulsed neutron beams are scattered off of the samples in various beam-lines. In the Sequoia Beam-line, a large area, position sensitive detector collects the scattered neutrons. Energy analysis comes from time-of-flight, momentum from the combination of energy and angle in the position sensitive detector. Thus, the data collection is four dimensional: energy and three components of momentum. The sample can also be rotated about a vertical axis, permitting different angles of incidence. Data analysis involves summing over various angles and energies, providing cuts through the multi-dimensional data space, to permit 2-D and 3-D plots. An example is shown Figure 1. This data can then be symmetrized, an example of which is shown Figure 2.

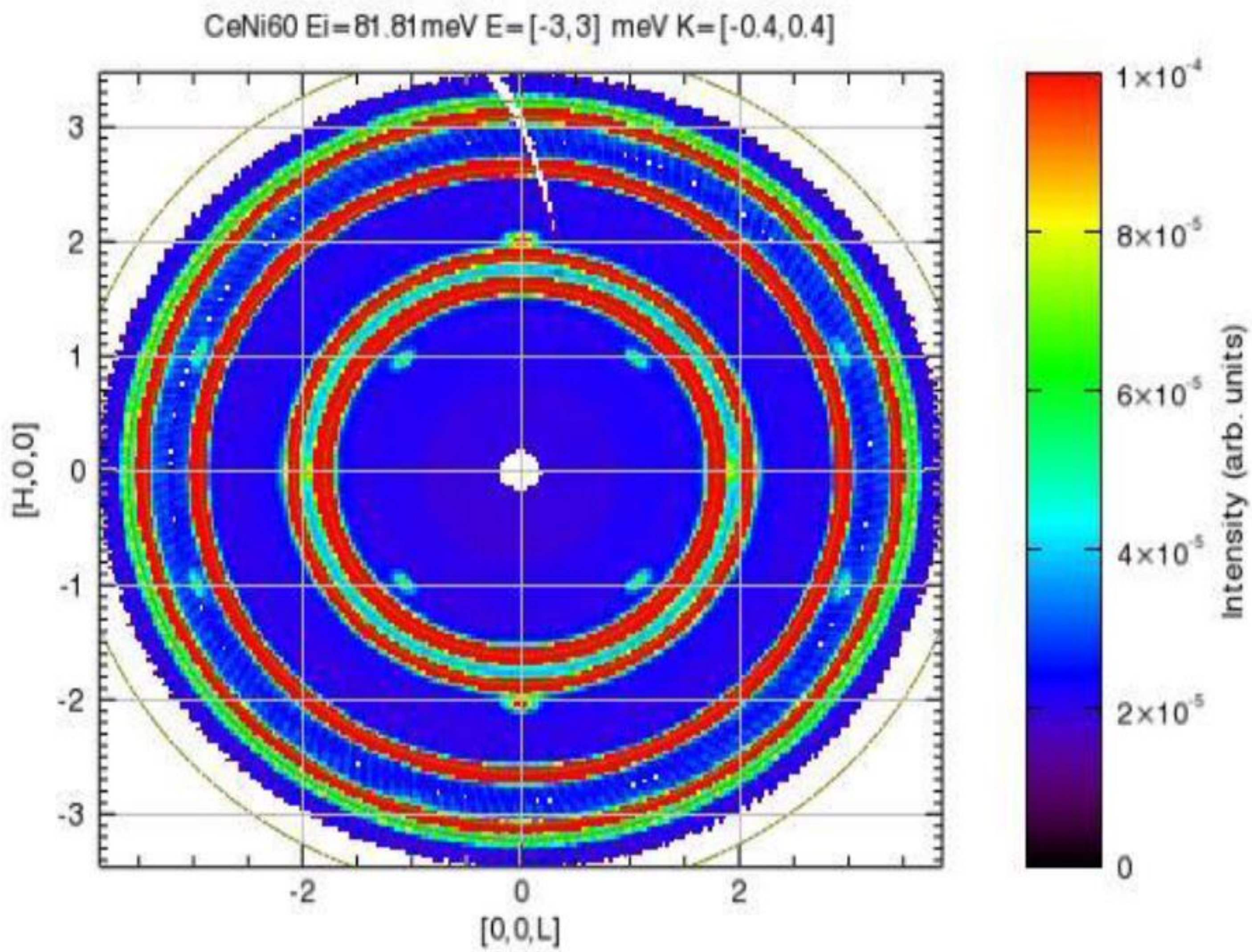

Figure 2: Scattering Data. The bright red and green rings are powder patterns from the polycrystalline Al of the pressure vessel. The individual spots are from the single crystal CeNi.

It is the energy loss that will provide a measure of the electron correlation. (Figure 3) After subtraction of the Al background, the data obtained at 400 Bar, 800 Bar and 2200 Bar suggest changes in the energy loss spectra. Further analysis is in progress. (All the data shown herein are at ambient pressure.)

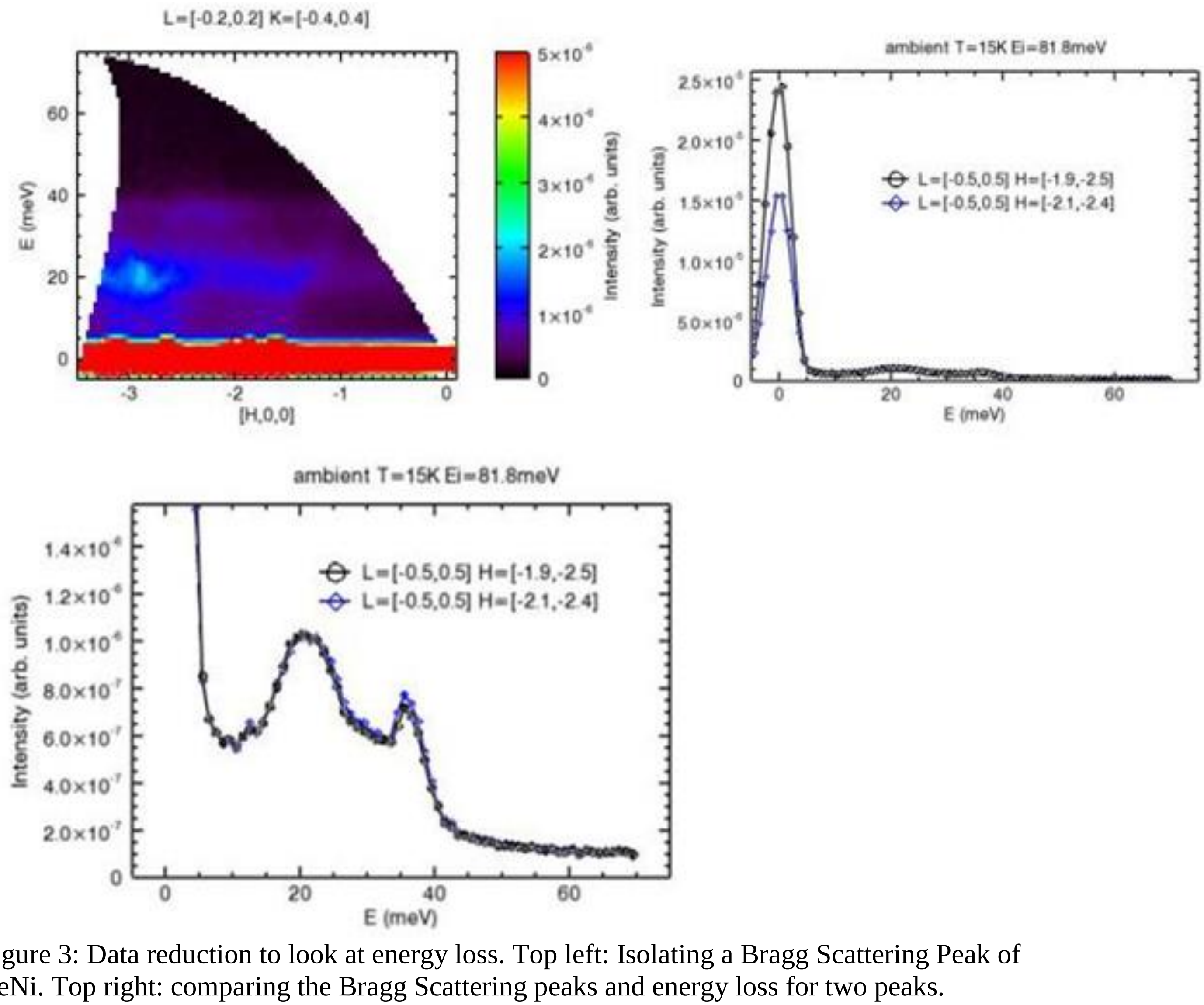

Figure 3: Data reduction to look at energy loss. Top left: Isolating a Bragg Scattering Peak of CeNi. Top right: comparing the Bragg Scattering peaks and energy loss for two peaks. Bottom: Blow-up of the comparison of the energy loss of two Bragg scattering peaks. This is raw data, without subtraction of the background from the Al pressure vessel.

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